OAK RIDGE NATIONAL LABORATORY

MANAGED BY UT-BATTELLE FOR THE DEPARTMENT OF ENERGY

P.O. Box 2008
Oak Ridge, TN 37831-6093
Phone: (865) 574-2480
E-Mail: hunnjd@ornl.gov

August 15, 2013

Dr. Paul Demkowicz
Idaho National Laboratory
P.O. Box 1625, 2525 North Freemont Avenue
Idaho Falls, Idaho 83415

SUBJECT: Completion of Level 2 Milestone Activity — Complete safety test report for ORNL-completed safety tests by September 15, 2013

Paul,

This letter formally documents successful completion of the Next Generation Nuclear Plant (NGNP) Level 2 Milestone (M2GR-13IN0501043), "Complete safety test report for ORNL-completed safety tests," due September 15, 2012. Work was performed under Memorandum Purchase Order MPO-83494, Statement of Work SOW-6904, Revision 6.

High-temperature safety tests were performed in the Core Conduction Cooldown Test Facility (CCCTF) at ORNL. Three Variant 3 fuel compacts from the AGR-1 irradiation experiment were individually subjected to maximum temperatures of 1600°C (Compact 4-1-2), 1700°C (Compact 4-4-3), or 1800°C (Compact 4-4-1) for a minimum of 300 hours each. Release of ^{85}Kr was monitored for indication of complete particle coating failure, and none was observed. Release of ^{134}Cs and ^{137}Cs was monitored for indication of SiC coating failure. Cesium release from Compact 4-4-3 was too low to measure. Cesium release from Compacts 4-1-2 and 4-4-1 was detected at levels typical for the release from one or two particles with breached SiC (the fact that these particles did not release krypton indicates one or more pyrocarbon layers remained intact). After deconsolidating Compacts 4-1-2 and 4-4-1, one particle with low cesium inventory was found in each compact. These particles were identified using the Irradiated Microsphere Gamma Analyzer (IMGA) to survey over 4000 particles from each compact. Additional post-safety test analysis of these particles is planned to investigate the cause of the cesium release. Other condensable fission products that escaped from the compacts were also collected throughout the CCCTF experiment. As expected, silver was the dominant fission product released through intact coating layers. Details of the CCCTF safety tests and post-safety test analyses will be documented in individual AGR-1 compact post-irradiation examination (PIE) reports. The attached safety test report summarizes the results of the three safety tests, including time-dependent and total fission product release results.

Sincerely,

John D. Hunn
Distinguished Research Staff
Oak Ridge National Laboratory

Attachment: ORNL/LTR-2013/290, *Safety Tests on Irradiated AGR-1 Compacts 4-1-2, 4-4-3, and 4-4-1*

SAFETY TESTS ON IRRADIATED AGR-1 COMPACTS 4-1-2, 4-4-3, AND 4-4-1

J.D. Hunn, R.N. Morris, C.A. Baldwin, and F.C. Montgomery

Summary

Post-irradiation examination (PIE) is in progress on coated particle fuel compacts from the first Advanced Gas Reactor (AGR) Fuel Development and Qualification irradiation experiment (AGR-1). The *AGR-1 Post-Irradiation Examination Plan* (INL/PLN-2828, Rev. 1) includes safety testing of the irradiated compacts in the Oak Ridge National Laboratory (ORNL) Core Conduction Cooldown Test Facility (CCCTF) and the Idaho National Laboratory (INL) Fuel Accident Condition Simulator (FACS) to evaluate the effect of elevated temperature on the fuel microstructure and fission product retention. Safety tests will involve heating compacts to maximum temperatures of 1600–1800°C for 100–500 hours (typically 300 hours), where 1600°C is the expected maximum temperature during a high-temperature gas-cooled reactor (HTGR) depressurization conduction cooldown event, and 1800°C is the expected temperature at which the performance of the SiC layer will begin to degrade.

ORNL completed the first AGR-1 post-irradiation compact safety test in FY2011, and four more safety tests were completed in FY2012. During the first three quarters of FY2013, three additional safety tests were completed. The first five safety tests at ORNL were performed on AGR-1 Baseline fuel from AGR-1 Irradiation Test Capsules 3 and 6. The three FY2013 safety tests discussed in this report all involved compacts from Capsule 4. Capsule 4 contained AGR-1 Variant 3 fuel, a coating variant which employed the addition of an argon diluent to the hydrogen and methyltrichlorosilane (MTS) used to deposit the SiC layer. The addition of argon results in a finer-grained SiC microstructure that is deemed to be desirable for optimum fuel performance; this coating variation has been adopted as the standard for AGR production-scale coating.

The first four ORNL safety tests were all performed at 1600°C, and the fifth was conducted at 1700°C. FY2013 safety tests involved maximum temperatures of 1600°C, 1700°C, and 1800°C. The standard heating profile specified in each compact examination plan called for a controlled ramp of the furnace to maximum temperature, with soaks at 400°C and 1250°C, a hold at maximum temperature for 300 hours, and then a controlled ramp back down to room temperature. The CCCTF performed as designed, except for a single interruption during the 1700°C furnace test on Compact 4-4-3 when an over-temperature warning due to insufficient cooling water flow to the gate valve thermal shield assembly caused the furnace power to shut down during the first 1700°C cup change; the cooling water supply issue was quickly corrected and the run was resumed.

During the test period, condensable fission products (metallic elements) were collected by the CCCTF in-furnace cold finger and gaseous fission products were collected from the helium sweep gas as it passed through liquid nitrogen-cooled cold traps. Deposition cups attached to the in-furnace cold finger were periodically removed and analyzed by gamma spectrometry and the sweep gas traps were constantly monitored for gamma activity throughout each run. Overall, the coated particle fuel continued to retain fission products generated during the three-year irradiation test, as designed. No significant gaseous ^{85}Kr release was detected by the sweep gas analysis system. Release of an appreciable amount of ^{85}Kr (typically more than half a particle's inventory) would be indicative of simultaneous failure of all three gas-tight outer layers of the tristructural isotropic (TRISO) coating, and is the primary indicator for catastrophic particle failure during the heating test. However, some radioactive fission products were released from the compacts during the tests. The main fission product detected on the deposition cups was ^{110m}Ag; lesser amounts of ^{154}Eu, ^{155}Eu, and ^{90}Sr were also detected. During the Compact 4-1-2 and Compact 4-4-1 safety tests, ^{134}Cs and ^{137}Cs were detected at levels typical for the release from one or two particles with breached SiC; the fact that these particles did not release krypton indicates one or more pyrocarbon layers remained intact.

After completion of each safety test, additional analysis was performed on the deposition cups and CCCTF furnace internals (graphite fuel holder, tantalum furnace liner, and tantalum gas inlet line) to measure the activity from the radioactive fission products that had been transferred from the compact to these various furnace components. An average deposition cup collection efficiency was determined for each fission product by measuring the total release from the compact (summing the inventories measured on the deposition cups, graphite holder, and tantalum furnace internals), and calculating the fraction of the total that went to the cups. The average collection efficiency was used to adjust the time-dependent deposition cup data to estimate the total time-dependent fission product release from the compact.

Post-safety test analysis of the furnace internals indicated high deposition cup collection efficiencies for silver and cesium, but only moderate efficiency for strontium and europium. Silver and cesium are both volatile at 1600–1800°C; this explains why most of what was released condensed on the cooled deposition cups and very little remained in the graphite holder or tantalum components. Europium and strontium easily form carbides, and typically half of what was released from the compact remained in the graphite holder and tantalum can. Compared to silver and cesium, the smaller fraction of europium and strontium on the deposition cups resulted in larger uncertainty in the total time-dependent fission product release values.

Compact 4-1-2 Safety Test

AGR-1 Compact 4-1-2 was irradiated at the INL Advanced Test Reactor (ATR) to an average calculated burnup of 17.3% fissions per initial metal atom (FIMA) and an average calculated fast fluence of $3.7 \cdot 10^{25}$ n/m^2, at a calculated volume-averaged and time-averaged temperature of approximately 1042°C. The Compact 4-1-2 safety test began on October 3, 2012, and ended on October 17, 2012. The compact was taken to a maximum test temperature of 1600°C and held there for 300 hours. The overall time-dependent results of the Compact 4-1-2 furnace test are shown in Figure 1. The plotted fractional release values are the cumulative amount of each fission product collected on the deposition cups, adjusted for radioactive decay and average collection efficiency, and divided by the predicted compact inventory for each fission product at the end of the irradiation. Very good agreement was obtained between the two isotopes of cesium and the two isotopes of europium.

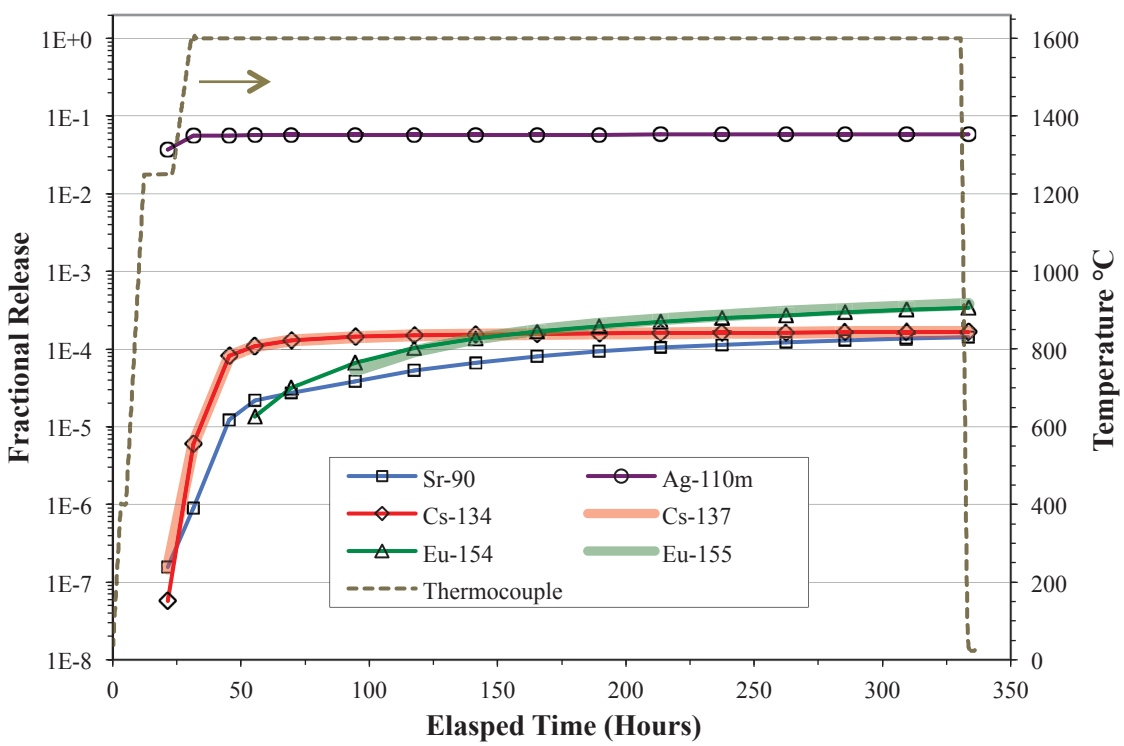

Figure 1. Release of fission products from Compact 4-1-2 during safety testing to 1600°C.

Table 1 lists the total furnace test releases for each detected fission product. A significant fraction of the calculated ^{110m}Ag inventory was detected on the deposition cups. Similar to previous safety tests, silver release dropped off after the initial rise to 1600°C. Constant exposure at 1600°C for 300 hours did not result in any significant additional silver collection. This indicates that silver is probably coming from what was trapped in the compact outside of intact SiC at the conclusion of the irradiation test. Cesium was also released early in the safety test, but it is likely this was related to individual particles, as discussed below. Europium and strontium releases were gradual, as previously observed; this is thought to be due to slow transport of these carbide-forming elements through the graphite in the compact matrix and the CCCTF fuel holder.

Table 1. Cumulative fission product release from Compact 4-1-2 safety test

Isotope	Fraction of Calculated Compact Inventory Released	Equivalent Particle Inventory
^{85}Kr	< 1E-06	< 0.004
^{90}Sr	1.43E-04	0.59
^{110m}Ag	5.80E-02	239
^{134}Cs	1.66E-04	0.69
^{137}Cs	1.66E-04	0.69
^{154}Eu	3.39E-04	1.40
^{155}Eu	3.79E-04	1.56

Note: One particle is equivalent to a compact inventory fraction of 2.42E-04.

The amount of cesium released from the compact was equivalent to the cesium expected to be released from one or two particles with a through-wall defect or failure of the SiC layer. After completion of the safety test, Compact 4-1-2 was electrolytically deconsolidated to recover the individual TRISO particles. Each particle was surveyed using the ORNL Irradiated Microsphere Gamma Analyzer (IMGA) to detect any particle with unusually low cesium or cerium content. One particle was identified with a very low cesium inventory that would account for the amount of cesium released during the safety test. As part of the post-safety test PIE, this particle will be examined using x-ray tomography and materialographic cross sectioning to investigate the causes for the elevated levels of fission product release. The fact that no ^{85}Kr was detected during the CCCTF test indicates that one or both of the pyrocarbon layers on this particle remained intact until the end of the safety test. The fact that negligible cesium was detected on the Capsule 4 components indicates that this abnormal particle did not release cesium during the irradiation test; this suggests that an open pathway through the SiC did not exist until the particle was heated to 1600°C during the post-irradiation safety test.

Compact 4-4-3 Heating Test

AGR-1 Compact 4-4-3 was irradiated at the INL ATR to an average calculated burnup of 18.8% FIMA and an average calculated fast fluence of $4.1 \cdot 10^{25}$ n/m^2, at a calculated volume-averaged and time-averaged temperature of approximately 1059°C. The Compact 4-4-3 safety test began on April 2, 2013, and ended on April 17, 2013. The compact was taken to a maximum test temperature of 1700°C and held there for 300 hours; this was the second 1700°C AGR-1 safety test (AGR-1 Baseline Compact 3-3-1 was tested at 1700°C in FY2012). The final heating profile and overall time-dependent release results of the Compact 4-4-3 furnace test are shown in Figure 2. The interruption at the start of the test due to insufficient cooling water is reflected in the drop from 1700°C back to room temperature that occurred 33 hours into the test. The two europium isotopes were in excellent agreement, and europium and strontium showed the typical trend of gradual release throughout the test. Because the ^{134}Cs and ^{137}Cs activities on various components was very low compared to possible background contamination levels, the cesium release could not be quantitatively determined.

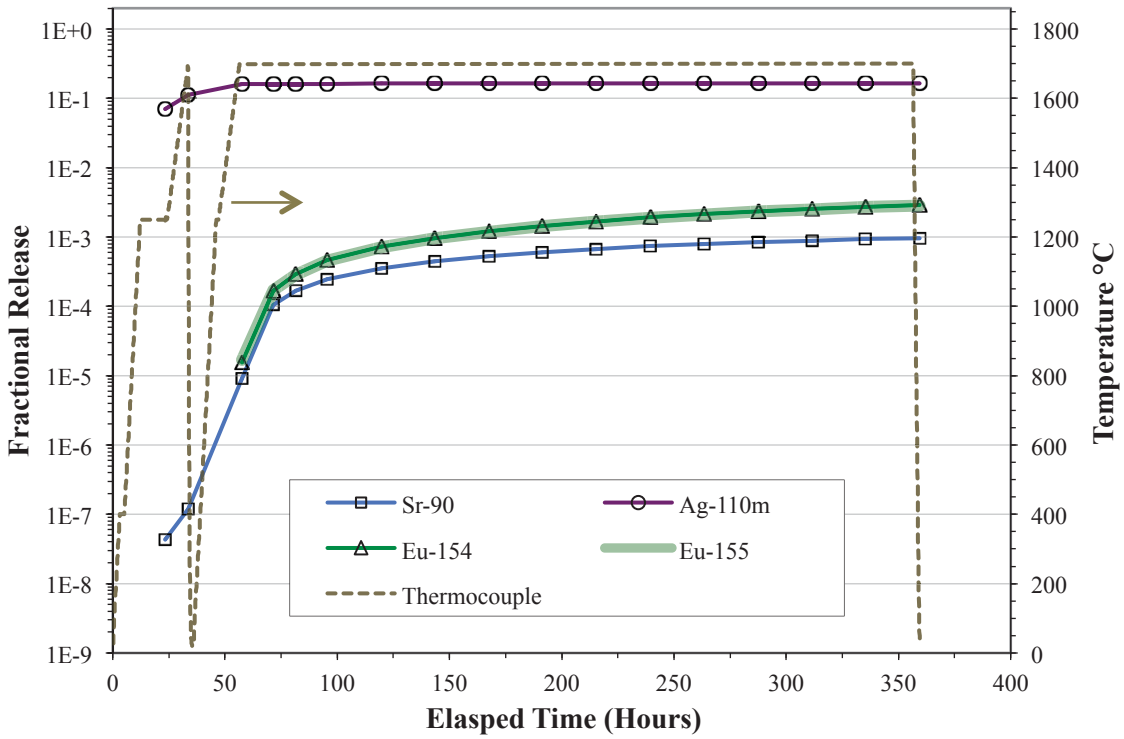

Figure 2. Release of fission products from Compact 4-4-3 during safety testing to 1700°C.

Table 2 lists the total furnace test releases for each detected fission product. Again, a significant fraction of the irradiated compact inventory of ^{110m}Ag was released in the early stages of the safety test. After the test was restarted, constant exposure at 1700°C for 300 hours did not result in any significant additional silver release. This is similar to the results of the Compact 4-1-2 safety test, and consistent with the conclusion that the silver that comes out during a normal safety test is probably already outside of intact SiC layers. However, Figure 2 shows that a measurable increase in ^{110m}Ag release of approximately 45% was detected when Compact 4-4-3 was heated to 1700°C a second time. For a typical safety test, 95–99% of the ^{110m}Ag is collected on the first cup removed after the compact reaches the test temperature. The additional ^{110m}Ag release from Compact 4-4-3 that was detected immediately after the compact was reheated to 1700°C may be related to a thermal cycling effect observed during previous safety tests. For instance, in the Compact 3-3-2 safety test, additional ^{110m}Ag was released every time the compact was cooled and reheated to 1600°C. Further study will be required to verify this effect and determine the source of any additional silver that may be released by thermal cycling.

Table 2. Cumulative fission product release from Compact 4-4-3 safety test

Isotope	Fraction of Calculated Compact Inventory Released	Equivalent Particle Inventory
^{85}Kr	< 1E-06	< 4E-03
^{90}Sr	9.67E-04	3.99
^{110m}Ag	1.66E-01	683
^{134}Cs	< 1E-06	< 4E-03
^{137}Cs	< 1E-06	< 4E-03
^{154}Eu	2.90E-03	12.0
^{155}Eu	2.85E-03	11.8

Note: One particle is equivalent to a compact inventory fraction of 2.42E-04.

Compact 4-4-1 Heating Test

AGR-1 Compact 4-4-1 was irradiated at the INL ATR to an average calculated burnup of 18.8% FIMA and an average calculated fast fluence of $4.0 \cdot 10^{25}$ n/m^2, at a calculated volume-averaged and time-averaged temperature of approximately 1057°C. The Compact 4-4-1 safety test began on February 18, 2013, and ended on March 4, 2013. The compact was taken to a maximum test temperature of 1800°C and held there for 300 hours; this was the first 1800°C AGR-1 safety test. The final heating profile and overall time-dependent results of the Compact 4-4-1 furnace test are shown in Figure 3. Again, excellent agreement was obtained for the two isotopes of cesium and europium. A very low level of ^{85}Kr was detected in the sweep gas traps after about 100 hours at temperature (the minor fluctuation in the plotted ^{85}Kr data is due to the fact that the values are close to the detection limit).

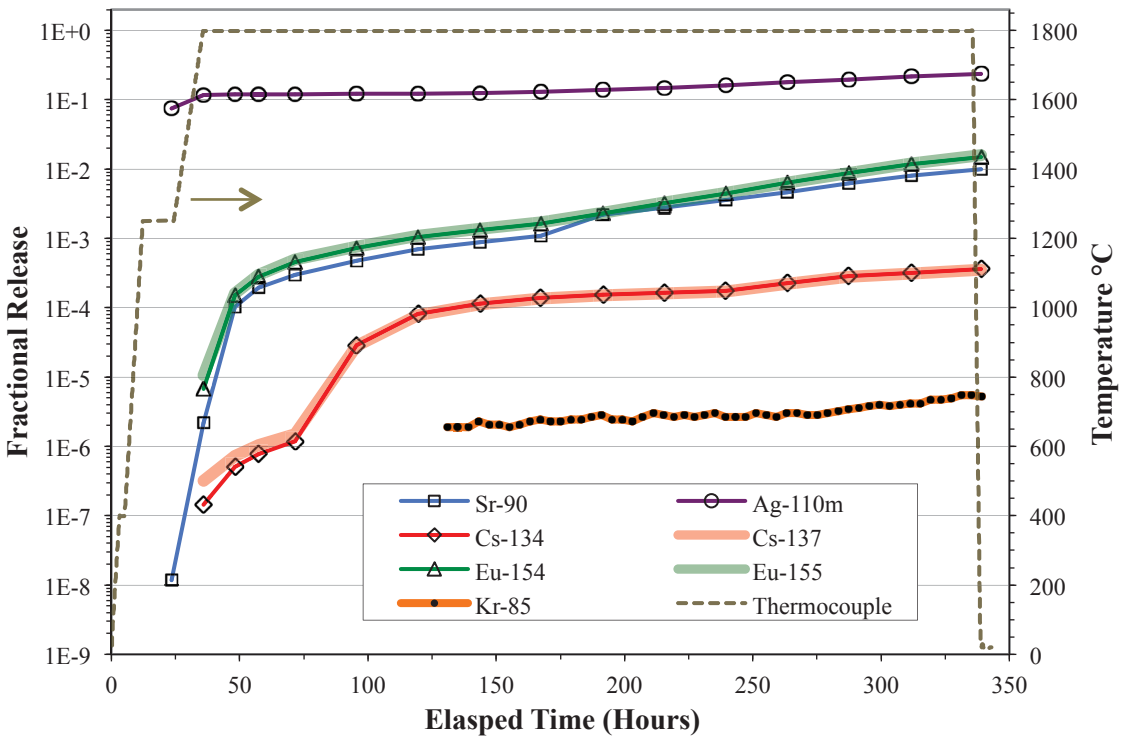

Figure 3. Release of fission products from Compact 4-4-1 during safety testing to 1800°C.

Table 3 lists the total releases for each detected fission product. Figure 3 shows that an increase in the cesium release was detected approximately 96 hours into the test (after ~60 hours at 1800°C), and a second increase in cesium release was detected approximately 263 hours into the test (after ~203 hours at 1800°C). The amount of cesium released after each detected increase suggests that two particles were involved, one for each event. Compact 4-4-1 was electrolytically deconsolidated to recover the individual TRISO particles, and each particle was surveyed with IMGA. Only one particle was identified with abnormally low cesium that might be related to the cesium release detected during the safety test. However, when this particle was transferred from a storage bottle to a tray for imaging prior to IMGA survey (Figure 4), the outer pyrocarbon (OPyC) layer was missing and the inner pyrocarbon (IPyC) and SiC layers separated into two hemispheres and broke away from the buffer-coated kernel. The gamma activity in the buffer-coated kernel and the two pieces of IPyC/SiC showed that the ^{137}Ce, ^{144}Ce and ^{154}Eu inventories were all very low, and no ^{110m}Ag activity could be detected. As was discussed regarding the cesium release in the Compact 4-1-2 safety test, because very little ^{85}Kr was detected during the Compact 4-4-1 safety test and there was no indication of cesium release in Capsule 4 during irradiation, it is apparent that the SiC layer of this particle failed after Compact 4-4-1 was heated to 1800°C and one or both of the pyrocarbon layers on the particle remained intact until the end of the safety test. The compact deconsolidation and subsequent handling prior to IMGA apparently caused additional damage to this

particle, and possibly to the second suspected cesium-releasing particle that was never found. The added stress of the 1800°C test may have been a contributing factor to these particles not surviving the standard electrolytic deconsolidation, acid leaching, and sieving process typically used to recover particles for IMGA survey. As part of the post-safety test PIE, the buffer-coated kernel and IPyC/SiC fragments from the particle with low cesium will be examined using x-ray tomography, and possibly materialographic cross sectioning, to further understand why this particle did not survive.

Table 3. Cumulative fission product release from Compact 4-4-1 safety test

Isotope	Fraction of Calculated Compact Inventory Released	Equivalent Particle Inventory
^{85}Kr	5.30E-06	0.02
^{90}Sr	9.99E-03	41.2
^{110m}Ag	2.34E-01	967
^{134}Cs	3.62E-04	1.49
^{137}Cs	3.53E-04	1.46
^{154}Eu	1.51E-02	62.1
^{155}Eu	1.58E-02	65.0

Note: One particle is equivalent to a compact inventory fraction of 2.42E-04.

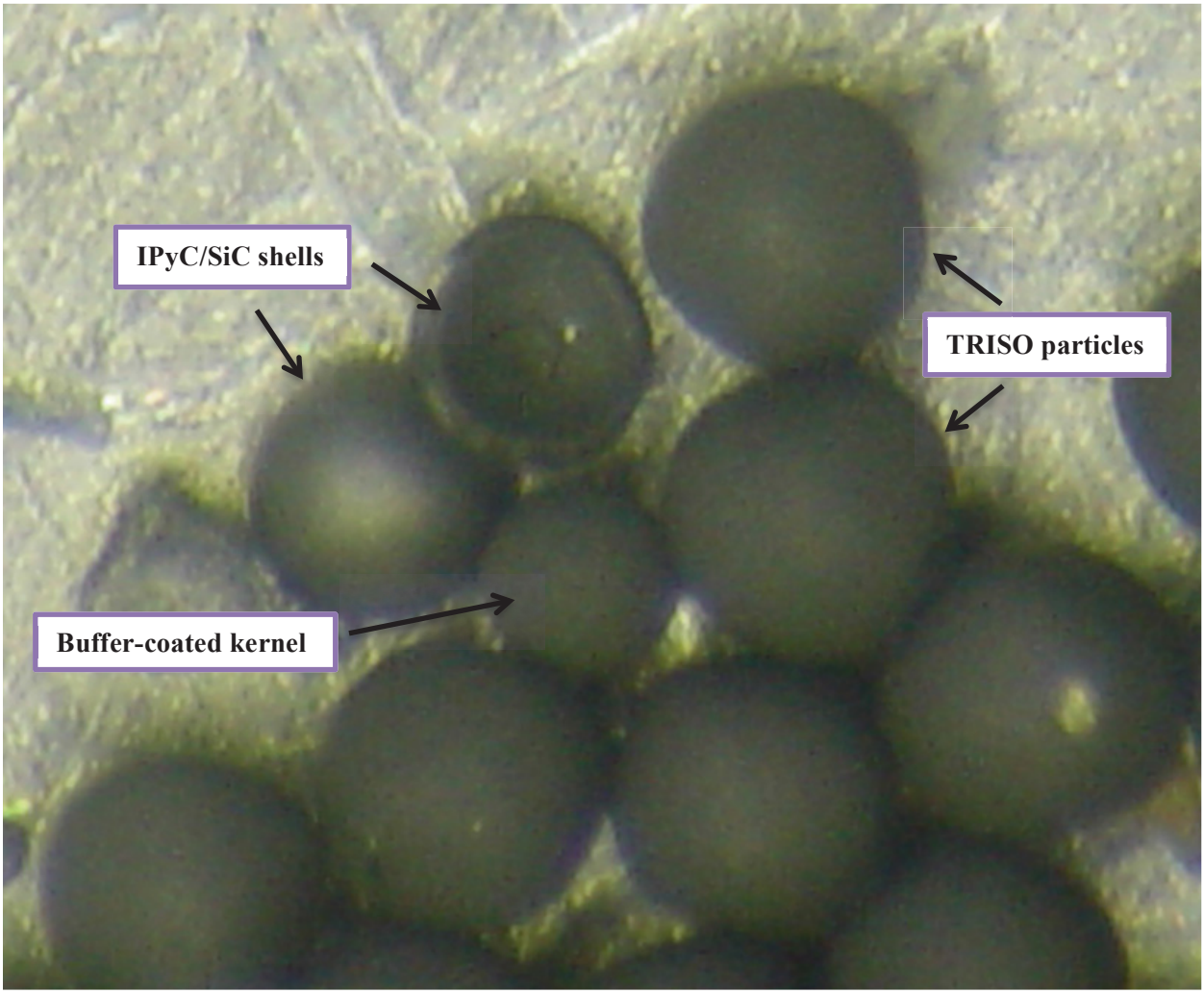

Figure 4. Defective particle recovered from Compact 4-4-1 after safety testing to 1800°C.

Figure 3 shows a noticeable increase in the ^{110m}Ag release after about 100 hours at 1800°C. This is in contrast to safety tests performed at lower temperatures, where an initial ^{110m}Ag release typically occurs as the compact is brought up to the test temperature, followed by no further significant release. In the 1800°C safety test of Compact 4-4-1, the ^{110m}Ag release rate continued to increase as the test progressed beyond 100 hours. The amount of ^{110m}Ag released during the later half of the test was roughly equivalent to what was initially released as the compact was heated to 1800°C (about 10% of the irradiated compact inventory in both cases). Whereas the initial release is thought to come from silver released during irradiation, this second release appears to be from diffusion through the SiC. Similar behavior was observed for europium and strontium.

Congruent with the additional release of silver, europium, and strontium, a small amount of ^{85}Kr was detected in the sweep gas trap (2% of one particle's inventory). The amount of ^{85}Kr released was not enough to have come from a particle with a TRISO-coating failure (typically >50% of one particle's inventory is released when a particle fails at safety test temperatures). The source of the ^{85}Kr release is not known. It is unlikely that krypton is passing through intact SiC, although the timing suggests that it could be somehow related to the additional releases of various metallic fission products throughout the latter portion of the test. It is more likely that the krypton release is coming from the one or two particles that released cesium. In the 1700°C Compact 3-3-1 safety test, two particles were found that released cesium during the test due to through-layer defects in the SiC layer; this test also showed a small ^{85}Kr release (about 3% of one particle's inventory).

Conclusion

The CCCTF performed well in FY13; three tests are reported here, and two additional FY13 safety tests performed on AGR-1 Variant 1 fuel will be discussed in a later report. As originally designed, the cooled-deposition cups have a high efficiency for collection of silver and cesium, and the sweep gas trap system has a good sensitivity to release of ^{85}Kr. Time-dependent monitoring of europium and strontium (not originally part of the CCCTF design) includes a larger uncertainty, due to the fact that these elements do not readily release from the graphite and tantalum furnace internals; analysis of the CCCTF furnace internals at the end of each test was important for accurate assessment of the total cumulative release of these elements. Very good agreement is being obtained between observations made during the safety tests and post-safety test PIE, such as analysis by leach-burn-leach (LBL) and IMGA. In addition, the extraction of individual particles with abnormally high fission product release is being pursued whenever possible, and this activity is important for determination of failure mechanisms.

Cesium continues to be retained well when compacts are heated during post-irradiation safety testing, even up to 1800°C, except when particles have through-wall defects in the SiC layer. The lack of significant ^{85}Kr release into the sweep gas indicates the absence of any catastrophic failures in the TRISO coatings as a whole, with the pyrocarbon layers serving as a secondary line of defense for those few particles with defective or failed SiC. Significant quantities of silver and europium were released from the compacts during the safety tests, far beyond what can be attributed to through-wall defects in the SiC layer. At 1600 and 1700°C, silver appears to come out of the compact immediately upon heating, but then no further release occurs unless the temperature is cycled; collection of europium and strontium on the deposition cups occurs over a longer time period. The ^{110m}Ag detected at the beginning of the 1600°C and 1700°C tests, and the europium and strontium collected on the deposition cups throughout these tests, are probably coming from the compact matrix, as opposed to transport through intact SiC layers. Silver and europium are known to escape through intact SiC during irradiation, and the quantities being detected during the safety tests are in the range of what has been detected by LBL in the matrix of as-irradiated compacts not subjected to safety testing. At 1800°C, additional silver, as well as europium and strontium, appear to be released after ~100 hours at temperature, and the cumulative release of all these fission products is higher at this higher temperature. Post-safety test PIE should be performed to investigate the possibility that these delayed releases may be associated with thermal diffusion and breakthrough.

Palladium release has been detected on the deposition cups and other furnace internals, but results are not reported here because additional work is required to determine quantitative release fractions. The ^{105}Pd

isotope has been used to track palladium in compacts analyzed by LBL. However, the copper coating on the CCCTF deposition cups interferes with this analysis ($^{65}Cu^{40}Ar^{+}$ forms in the mass spectrometer plasma). Other isotopes of palladium can be measured (e.g., ^{104}Pd, ^{106}Pd, and ^{108}Pd), but interferences with ruthenium and cadmium must be considered. Mass spectrometer data on these additional isotopes are being extracted from various analyses to determine the best approach for quantifying palladium release during CCCTF safety testing.

Acknowledgments

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